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CR 151327

FINAL RESEARCH REPORT TO THE
NASA LYNDON B. JOHNSON SPACE CENTER

MEASUREMENTS OF RADON CONCENTRATIONS
IN THE LUNAR ATMOSPHERE

NAS 9-13268

March 31, 1977

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(NASA-CR-151327) MEASUREMENTS OF RADON
CONCENTRATIONS IN THE LUNAR ATMOSPHERE
(Battelle Pacific Northwest Labs.) 17 p
MC A02/MF A01 CSCI 03E

N77-22035

Unclas
G3/91 24461

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ABSTRACT

The radon concentrations in the lunar atmosphere have been determined by measuring the ^{210}Po progeny activity in artifacts returned from the moon. Experiments performed on a section of the polished aluminum strut from Surveyor 3 yield an average lunar radon concentration of $(3.8 \pm 1.1) \cdot 10^{-3}$ disintegrations $\text{cm}^{-2} \text{ sec}^{-1}$ during its 944 day exposure at Oceanus Procellarum. Data obtained from the Apollo 16 Cosmic Ray Detector Experiment Teflon thermal shield yield a radon concentration of $(64 \pm 12) \cdot 10^{-3}$ disintegrations $\text{cm}^{-2} \text{ sec}^{-1}$ at the time of that mission in the Descartes area. These results are compared with other values of the lunar radon concentration obtained at different times and different locations and by various techniques. Possible sources and release mechanisms compatible with all of the data are discussed.

An experimental procedure to determine the relative retention coefficients of various types of material for radon progeny in a simulated lunar environment is described. The results of several experiments are given, and their effect on lunar radon progeny measurements is discussed.

An analytical procedure is given for the analysis of a Teflon matrix for trace constituents.

INTRODUCTION

The episodic evolution of radiogenic gases from the interior of the moon into the lunar atmosphere has been fairly conclusively demonstrated. Evidence for this phenomenon has been gathered from earth-based observations, from direct measuring instruments placed on the moon during the Surveyor and Apollo missions, from instrumental measurements made by lunar orbiting spacecraft, and from analysis of returned lunar materials and artifacts. A fairly concise summary of many possible sources and release mechanisms for this episodic evolution of radiogenic gas is given by Friesen⁽¹⁾.

The objectives of this research effort were to determine the equilibrium activity of ^{222}Rn in the lunar atmosphere at the Apollo landing sites by measuring the entrapped radon decay products in a polished tube section from the Surveyor 3 spacecraft, in the Teflon thermal shield from the Apollo 16 Cosmic Ray Detector Experiment, in the Solar Wind Composition Experiment foils, and in the Kapton thermal coatings of the command modules which orbited the moon. In addition, the relative retention coefficients of each of these materials was determined in the laboratory in a simulated lunar environment to see if there might be significant differences in radon progeny collection efficiency. Any affinity of the materials for gaseous radon would result in an increase in the apparent radon concentration in the near vicinity of the material during exposure. The selective rejection of radon progeny after incidence on the material would be manifested in a lower apparent radon concentration.

The results of this program have been examined in light of proposed sources and episodic release mechanisms and compared with other data obtained by different techniques.

EXPERIMENTAL PROCEDURES

Radon atoms bounce along the moon in ballistic trajectories since their thermal energy is insufficient to provide escape velocity. As these atoms undergo radioactive decay, relatively large amounts of kinetic energy are imparted to the ^{218}Po daughters. Only slightly more than half of these daughters will strike the moon, since most of those recoiling away from the moon will escape. Those recoils which strike an object exposed to the lunar environment embed themselves and are trapped. Again, almost half of the trapped atoms are lost from the material by subsequent alpha decays to ^{214}Pb and ^{210}Pb , but this loss is very nearly compensated by the gain from the same decays on the surface of the moon.

Range-energy calculations indicate that a two-sided planar material, such as the SWC foil, will maintain 48% of the radon decays in the form of ^{210}Pb , assuming no enhancement or loss of radon progeny due to effects other than radioactive recoil. A one-sided planar material in lunar orbit, such as the thermal coating on the command and service module, will effectively capture 25%. A one-sided planar material on the surface of the moon, such as the Cosmic Ray Detector Experiment Teflon thermal shield, will have a capture efficiency of 29%, and a cylindrical object, such as the Surveyor 3 strut, will have the same efficiency as a two-sided planar material of the same surface area.

Polonium-210 is an easily measurable decay product of the 20.4 year half-life ^{210}Pb and can be used to determine the ^{210}Pb concentrations in the exposed material, and hence, the radon concentrations in the lunar atmosphere at the time of exposure. Unfortunately, the technique does not afford a simultaneous measurement of the ^{210}Po on the lunar surface at the same location; therefore, a precise determination of the degree of equilibrium between radon and polonium is not possible by this method.

Candidate materials for radon exposure measurements must meet several criteria including low intrinsic ^{210}Po content, availability of suitable blank material, and documented exposure to the lunar atmosphere. The SWC foils, the thermal coatings from the spacecraft and from the Apollo 16 Cosmic Ray Detector Experiment, and the polished aluminum strut from the Surveyor 3 spacecraft have proven to be suitable collectors.

A detailed description of the analytical procedure for determination of ^{210}Po including an evaluation of possible experimental problems associated with such ultra-low level radioactivity measurements has been published elsewhere⁽²⁾ and will not be repeated here.

Due to the inherent insolubility of Teflon, a special dissolution procedure had to be developed for analysis of the Apollo 16 Cosmic Ray Detector Experiment Teflon thermal coating. In this procedure, the sample, a 6 x 8 cm piece of Dupont type A FEP Teflon 0.051mm thick and coated on one side with $\sim 250\text{\AA}$ of inconel and $\sim 1500\text{\AA}$ of silver, is ashed for two hours in a stainless steel bomb at 550°C . Utilizing a nitrate-peroxide oxidizing system, a "sandwich" consisting of layers of NaNO_3 , Na_2O_2 , teflon, Na_2O_2 , NaNO_3 is placed in a 2 cm diameter by 7 cm deep cavity and heated to fuse the Teflon. After cooling the melt is dissolved in warm dilute nitric acid followed by the usual chemical separations.⁽²⁾

A special procedure for analysis of the Surveyor 3 aluminum strut was also required due to the relatively high intrinsic ^{210}Po content of the aluminum. A surface etch technique was used to remove a minimum quantity of aluminum from the strut commensurate with removing 100% of all embedded ^{222}Rn progeny. In this manner, the background from intrinsic ^{210}Po in the aluminum could be minimized. The aluminum tubing was etched in 1N HCl for two minutes, and weight loss was used to determine the amount of surface erosion. Analyses of

the acid solutions by the standard procedure⁽²⁾ yielded the specific activity of ^{210}Po in the strut and blank material.

To guarantee the reliability of measuring ^{210}Po concentrations in lunar artifacts as a method of estimating the radon content of the lunar atmosphere, it is necessary to establish the radon progeny retention coefficients of the materials. This is to determine whether the materials possess either a "getter" quality for radon which might artificially enhance the apparent radon concentration to which it was exposed or a spontaneous rejection of radon progeny which would have the opposite effect. To evaluate these effects in the laboratory, it was necessary to simulate lunar exposure conditions while still maintaining a workable time frame.

Blank material of each lunar artifact plus two additional reference materials were exposed to relatively high concentrations of radon gas at sufficiently low pressure to simulate the lunar deposition conditions for radon and its daughter products. The quantities of ^{218}Po , ^{214}Pb , ^{214}Bi , and ^{214}Po were determined in the exposed foils using Thomas'⁽³⁾ method of interpreting time variation of alpha particle decay after exposure. Radon-222 was measured directly in one experiment utilizing a solid state alpha energy spectrometer.

The exposure apparatus consisted of a modified glass enclosed vacuum coater which had a cover 60 cm high and 44 cm in diameter mounted on a stainless steel base. Evacuation was by combination of a mechanical pump, an oil diffusion pump, and a liquid nitrogen cooled sorption pump which furnished chamber pressures as low as $3 \cdot 10^{-7}$ torr.

Rectangular pieces, 3.81 cm x 5.08 cm, of each lunar artifact material, silicon monoxide coated aluminized kapton spacecraft thermal shield, aluminum oxide coated aluminum solar Wind Composition foil, silver and inconel coated type "A" FEP Teflon Cosmic Ray Detector Experiment thermal shield, and

aluminum Surveyor 3 strut, plus samples of uncoated type "A" FEP Teflon and polyethylene, were located symmetrically in the vacuum chamber. A known charge of ^{222}Rn sealed in a 24 karat gold ampoule 3.5 mm long and 0.75 mm outside diameter was placed in a guillotine located in the vacuum chamber and operated by a mechanical feedthrough. Confinement, leakage, and dispersion of the radon gas and its daughter products was monitored by scintillation gamma-ray spectrometry from the outside of the vacuum chamber. After evacuation, the chamber was isolated, and the gold ampoule severed in the guillotine to start the simulated lunar exposure. Pressure was monitored by ionization and thermocouple gauges. The simulated exposure was ended by bleeding several torr of room air into the chamber. The system was again evacuated and "rinsed" a total of three times with room air before opening.

Samples were counted on six zinc sulfide type alpha particle scintillation counters. Controllable parameters, such as geometry of the foils, electrical grounding, alpha detectors, etc., were varied in several different experiments in order to identify and eliminate any systematic errors.

RESULTS

The results of the experiments to determine the relative radon progeny retention coefficients of the various materials are summarized in Table I. The data are normalized to the Cosmic Ray Detector Experiment Teflon thermal shield which was found experimentally to agree precisely with the calculated values. No significant systematic effect was observed due to exposure position, electrical isolation, or counting facility.

The materials with metalized surfaces exposed showed significantly higher retention of radon daughters than did the plastic materials. This behavior is not unexpected considering the ion sticking probabilities of the various materials. The probability of metal ions sticking onto metal surfaces is

TABLE I

Relative Activity of Alpha Active Radon
Progeny in Various Materials Exposed to a
Simulated Lunar Atmosphere Normalized to
Aluminum

<u>Material</u>	<u>Relative Activity</u>
1. FEP Teflon	0.93±0.05
2. Silicon monoxide coated aluminized Kapton spacecraft thermal coating	1.21±0.08
3. Inconel and silver coated FEP Teflon Cosmic Ray Detector Experiment thermal shield	1.00
4. Aluminum - Surveyor 3 strut	1.16
5. Aluminum oxide coated aluminum Solar Wind Composi- tion Experiment detector	1.26±0.15
6. Polyethylene	0.97±0.02

higher than onto plastic surfaces due to the nature of the binding forces involved. Adhesion to plastic surfaces is primarily by physisorption (Van der Waals) forces, whereas binding to metal surfaces is by much stronger chemisorption forces.

In the experiment to measure the relative radon progeny retention coefficients of lunar artifacts during exposure to the simulated lunar atmosphere, it was possible to resolve the alpha particle decay curves such that the individual concentrations of ^{210}Po , ^{214}Pb , ^{214}Bi , and ^{214}Po in each material could be determined. The materials with plastic surfaces exposed (foils 1, 3, and 6 in Table I) demonstrated the predicted rate of disequilibrium, while the metalized materials (2, 4, and 5) did not. That is, the absolute values of each individual progeny present in only the plastic materials agree with the predicted values based on recoil loss. Therefore, the lunar radon concentration data obtained from metalized materials^(4,5) needs to be adjusted for the unforeseen retention of radon progeny.

Table II summarizes some previously published results of lunar radon and/or polonium measurements along with the two new pieces of data determined in the course of this research. Although it was planned to also investigate the radon progeny content of the Kapton thermal shields from all lunar orbiting spacecraft, these artifacts were not made available for the research.

DISCUSSION

The radon concentration at the Surveyor 3 landing site in Oceanus Procellarum was determined to be $3.8 \cdot 10^{-3}$ disintegrations $\text{cm}^{-2} \text{sec}^{-1}$ from measurement of the radon progeny in a section of the polished aluminum strut returned by the Apollo 12 astronauts. This value is an average for the 944 days in 1967-1969 that the strut was exposed to the lunar atmosphere and is compatible with the upper limit of ^{210}Po activity measured on the returned Surveyor 3 camera visor

TABLE II

Lunar ^{210}Po and ^{222}Rn Activities in Units of
 10^{-3} Disintegrations $\text{cm}^{-2} \text{sec}^{-1}$

<u>Method or Device†</u>	<u>Time Base of Determination</u>	<u>Location</u>	<u>^{210}Po</u>	<u>^{222}Rn</u>	<u>Ref.</u>
Calculation	Pre 1966	All	1300		6
Explorer 35 OAS	1967-1968	All	<110	<380*	7
Surveyor 5 Spectrometer	1967-1968	Mare Tran- quillitatis	29±9	100±31*	8
Surveyor 6 Spectrometer	1967-1968	Sinus Medii	<21	<110*	8
Surveyor 7 Spectrometer	1967-1968	Tycho	<21	<110*	8
Surveyor 3 Visor	1967-1969	Oceanus Procellarum	<5	<17*	9
Surveyor 3 Strut	1967-1969	Oceanus Procellarum	1.09±0.31*	3.8±1.1	This work
Lunar Fines	Pre 1969	Mare Tran- quillitatis	<0.45	<1.6*	10
Apollo 12 SWC Foil	1969	Oceanus Procellarum	18.9±2.8*	65.4±9.6	4
Apollo 12 Kapton	1969	All	20.7±9.1*	71±31	4
Lunar Fines	Pre 1971	Fra Mauro	11*	37	11
Lunar Fines	Pre 1971	Hadley- Apennine	11*	37	11
Apollo 14 SWC Foil	1971	Fra Mauro	<5.5*	<19	5
Apollo 15 SWC Foil	1971	Hadley- Apennine	6.9±1.1*	23.9±3.6	5
Apollo 15 & 16 OAS	1971-1972	All	3.1-4.6	0.92-1.32	12,13
Apollo 15 & 16 OAS	1971-1972	Aristarchus (^{222}Rn) Mare Fecunditatis (^{210}Po)	16	10.1	12,14
Apollo 16 SWC Foil	1972	Descartes	7.3±1.5*	25.2±5.0	5
Apollo 16 CRDE Foil	1972	All (17%) Descartes (83%)	18.5±3.6*	64±12	This work

*Calculated from measured parent or daughter activity.

†OAS = orbiting alpha spectrometer; SWC = Solar Wind Composition; CRDE = Cosmic Ray Detector Experiment

by Economou and Turkevich⁽⁹⁾. However, it should be noted that both the camera visor and the polished tube section were covered with a deposit which may have hindered the deposition or adhesion of radon progeny. This would result in apparent radon concentrations which were lower than actually existed at the time of exposure.

The Surveyor 3 strut data can also be compared to the radon concentration measured by Brodzinski⁽⁴⁾ at the Apollo 12 landing site from the Solar Wind Composition foil exposed at the time of the landing. The radon concentration determined from the Surveyor 3 strut is a factor of 17 lower than that inferred from the SWC foil and may be due to real differences in the average radon emission during the 944 day exposure of the strut and the 18 hour exposure of the SWC foil, or to biased results because of the deposit on the surface of the strut, or a combination of both.

The radon concentration at the Apollo 16 landing site was determined to be $64 \cdot 10^{-3}$ disintegrations $\text{cm}^{-2} \text{sec}^{-1}$ from the progeny activity in the Cosmic Ray Detector Experiment Teflon thermal shield. This artifact received 17% of its effective exposure to the lunar atmosphere from orbit and 83% of the exposure at the landing site in Descartes. This data can be compared to the data of Brodzinski and Langford⁽⁵⁾ obtained from the Apollo 16 Solar Wind Composition foil which received a nearly identical exposure. Corrections have been applied to the data from the solar wind composition foils, the Surveyor 3 strut, and the spacecraft Kapton coating to compensate for their slightly increased retention efficiency for radon daughters and to the Cosmic Ray Detector Experiment Teflon thermal shield for the single surface collection ability of this material compared to the double surface of the Solar Wind Composition foil. The Cosmic Ray Detector Experiment foil was exposed to an apparent radon concentration more than twice as high as that observed by the Solar Wind Composition foil. No document-

able explanation for this discrepancy is immediately apparent. The Cosmic Ray Detector Experiment was deployed on a footpad of the LEM while the Solar Wind Composition foil was erected at some distance from the LEM. Perhaps the increased astronaut activity in the immediate vicinity of the Cosmic Ray Detector Experiment enhanced soil radon emanation sufficiently to account for an extremely high localized concentration.

The possibility of localized physical activity as a primary source of radon emanation can be supported further if it is assumed that the Surveyor 3 data is not biased due to the deposit on the artifact surfaces. The reported radon concentration measured by the Apollo 12 Solar Wind Composition foil is substantially higher than that measured by the Surveyor 3 artifacts and could also be attributed to local astronaut activity at the time of exposure, since, except for landing and recovery, the Surveyor 3 artifacts spent the vast majority of their exposure on a quiescent moon.

Evidence against this local disturbance enhancement hypothesis is equally available since the ^{210}Po activities in the Apollo 12 Solar Wind Composition foil and the Apollo 16 Cosmic Ray Detector Experiment foil, which were exposed to relatively large amounts of astronaut activity, are not as high as that obtained with the Surveyor 5 spectrometer measurements of Turkevich, et al.⁽⁷⁾, which had absolutely no physical disturbance in the vicinity at the time of data accumulation.

When all possible sources of adverse influence on lunar radon measurements are considered, it becomes apparent that a general approach of orbital spectrometry such as that used by Gorenstein, Bjorkholm, and Golub^(12, 13, 14) has distinct advantages. However, accumulation of statistically significant data by this technique over a large area of the moon with a relatively fine spatial resolution requires much more sensitivity than was available on the Apollo

missions. Increased orbital sensitivity could be achieved on missions such as that proposed for Lunar Polar Orbiter by incorporation of large area spectrometers. However, such sensitive data would be expected to only further support the well-documented and accepted phenomenon of variant lunar radon emission. All evidence indicates relatively large spatial and temporal variations and a delicate sensitivity to external disturbances such as astronaut activity, moonquakes, etc. Although the sources and release mechanisms of volatiles on the moon as postulated by Friesen⁽¹⁾ are supported by this evidence, it appears that even more localized and miniscule mechanisms may have a substantial influence.

ACKNOWLEDGMENTS

The authors are indebted to J. Geiss, F. Buehler, H. Cerruti, P. Eberhardt, J. Meister, and Ch. Filleux for furnishing and preparing the Solar Wind Composition Experiment materials. Similar gratitude is extended to R. Fleischer for making the Cosmic Ray Detector Experiment Teflon thermal shield material available to us. We also thank M. Duke, J. Annexstad, and W. Eichelman at the NASA Johnson Space Center for their extended and diligent efforts at obtaining samples and data for us.

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